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A sub-field scale critical source area index for legacy phosphorus management using high resolution data

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ABSTRACT

Diffuse phosphorus (P) mitigation in agricultural catchments should be targeted at critical source areas (CSAs) that consider source and transport factors. However, development of CSA identification needs to consider the mobilisation potential of legacy soil P sources at the field scale, and the control of (micro) topography on runoff generation and hydrological connectivity at the sub-field scale. To address these limitations, a 'next generation' sub-field scale CSA index is presented, which predicts the risk of dissolved P losses in runoff from legacy soil P. The GIS-based CSA Index integrates two factors; mobile soil P concentrations (water extractable P; WEP) and a hydrologically sensitive area (HSA) index. The HSA Index identifies runoff-generating-areas using high resolution LiDAR Digital Elevation Models (DEMs), a soil topographic index (STI) and information on flow sinks and effects on hydrological connectivity. The CSA Index was developed using four intensively monitored agricultural catchments (7.5–11 km^2) in Ireland with contrasting agri-environmental conditions. Field scale soil WEP concentrations were estimated using catchment and land use specific relationships with Morgan P concentrations. In-stream total reactive P (TRP) concentrations and discharge were measured sub-hourly at catchment outlet bankside analysers and gauging stations during winter closed periods for fertiliser spreading in 2009–14, and hydrograph/loadograph separation methods were used to estimate TRP loads and proportions from quickflow (surface runoff). A strong relationship between TRP concentrations in quickflow and soil WEP concentrations ($r^2 = 0.73$) was used to predict dissolved P concentrations in runoff at the field scale, which were then multiplied by the HSA Index to generate sub-field scale CSA Index maps. Evaluation of the tool showed a very strong relationship between the total CSA Index value within the HSA and the total TRP load in quickflow ($r^2 = 0.86$). Using a CSA Index threshold value of $\geq$0.5, the CSA approach identified 1.1–5.6% of catchment areas at highest risk of legacy soil P transfers, compared with 4.0–26.5% of catchment areas based on an existing approach that uses above agronomic optimum soil P status. The tool could be used to aid cost-effective targeting of sub-field scale mitigation measures and best management practices at delivery points of CSA pathways to reduce dissolved P losses from legacy P stores and support sustainable agricultural production.

1. Introduction

Diffuse phosphorus (P) losses from agricultural land to surface waters continue to be a major pollution issue worldwide, causing deterioration of water quality and impacts on ecosystem services (European Environment Agency, 2015; McDowell et al., 2015; Sharpley and Wang, 2014). As a result, mitigation measures are part of wide ranging and international environmental policies and

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legislation (Schoumans et al., 2014; McDowell and Nash, 2012). Catchment areas at greatest risk of P transfers are termed critical source areas (CSAs), where high concentrations of mobile P coincide with hydrologically sensitive areas (HSAs) which have high runoff potential (Pionke et al., 2000; Walter et al., 2000). CSAs must be accurately identified if mitigation measures and best management practices are to be targeted, cost-effective and successful in reducing P losses (Doody et al., 2012; Sharpley et al., 2011).

A number of P CSA Indices exist, which range in terms of the source, mobilisation and transport factors used, weightings, formulation, and whether they predict relative P loss risk or quantify P loads (Heckrath et al., 2008; Buczko and Kuchenbuch, 2007). In the USA, where CSA Indices are termed P Indices, concerns have been raised over inconsistencies, state-by-state variability, and the lack of calibration and evaluation using measured P loss data (Osmond et al., 2012; Nelson and Shober, 2012). Slow improvements in water quality following over twenty years of regulatory implementation also suggest that such CSA definitions are limited (Sharpley et al., 2011, 2012), leading to calls for refinements (Sharpley et al., 2013a).

CSA Indices currently use agronomic soil P tests as a source factor. Soils with high total P concentrations have historically received excessive manure or fertiliser P applications that outweighed crop requirements (Kleinman et al., 2011). However, agronomic soil P tests do not consider the mobilisation potential of this residual or 'legacy' soil total P to be released to runoff pathways, despite mobilisation being a fundamental component of the P transfer continuum (Haygarth et al., 2005). The natural affinity of soils to bind and immobilise P varies based on soil properties such as aluminium (Al), iron (Fe), calcium carbonate, clay, pH and organic matter (OM), and hence in some soils, legacy P (total P) is more vulnerable to desorption, solubilisation and transport in surface runoff (Daly et al., 2001, 2015; Maguire and Sims, 2002). Thus different soils can have the same amount of total P, but different amounts of available P, and vice-versa.

Environmental soil P tests such as water extractable P (WEP) (also known as water soluble P) are considered better at replicating the chemical interaction between soil P and runoff and are less affected by soil type, and hence are arguably better at predicting dissolved P concentrations in runoff (Torbert et al., 2002; Pote et al., 1999; Penn et al., 2006). Some CSA Indices already use the WEP test as a 'P source coefficient factor' to predict the mobilisation potential of fertiliser P in runoff (Kleinman et al., 2007; Shober and Sims, 2007). However, the WEP test has not yet been widely applied as a specific legacy soil P risk assessment; exceptions include Regan et al. (2010, 2014), Ulén et al. (2011), Djodjic and Bergström (2005), and Dodd et al. (2012).

Another limitation of conventional CSA definitions is the use of watercourse proximity as a proxy for runoff risk and P transport potential (Gburek et al., 2000; Srinivasan and Mcdowell, 2007). This is recognised as an extreme simplification of reality which does not account for the effects of (micro)topography on the generation, channelisation, convergence and hydrological

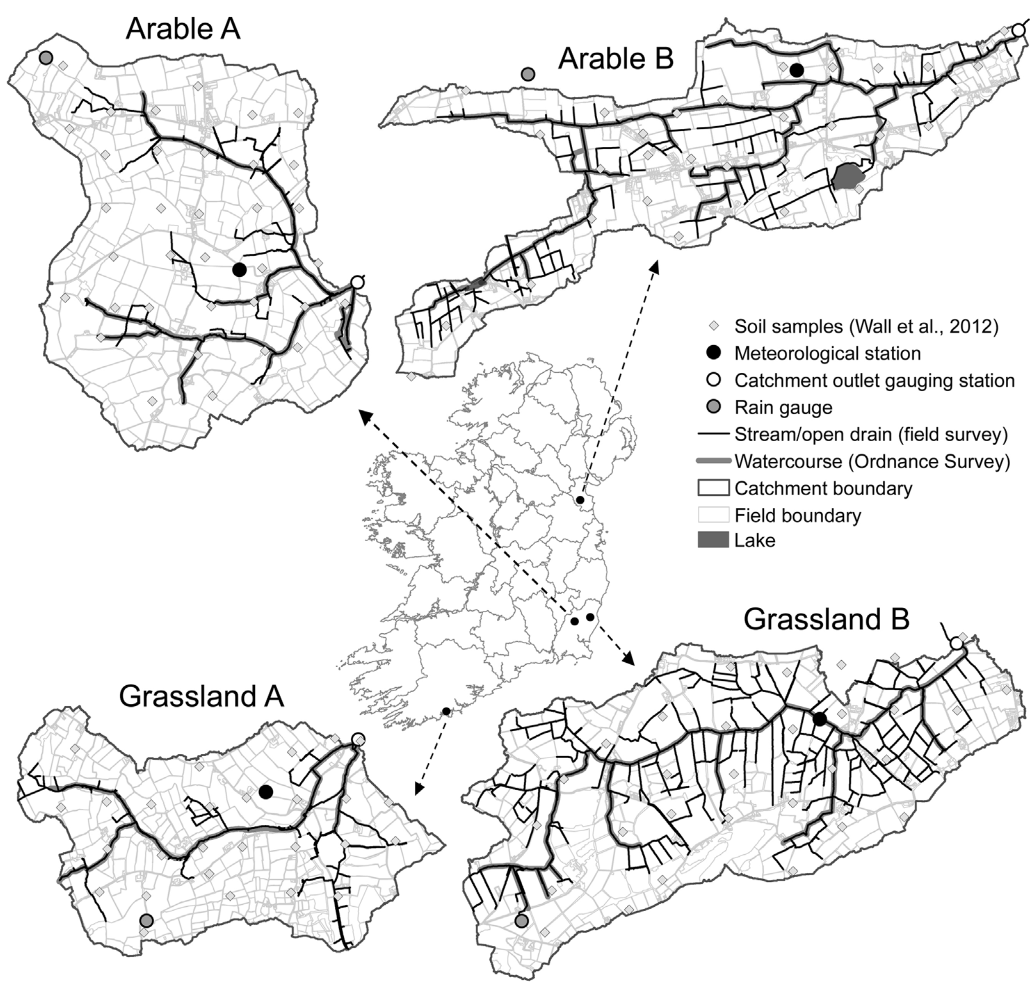

Fig. 1. Locations of the agricultural catchments in Ireland used in the study. Also indicated are the stream and drainage channel networks, catchment outlet gauging station, meteorological station, and locations of the spatially stratified soil samples taken by Wall et al. (2012).

connectivity of overland flow (Marjerison et al., 2011; Thomas et al., 2016, 2017; Lane et al., 2009). The approach therefore tends to overestimate HSA extents at the stream and underestimate extents upslope (Sharpley et al., 2013a; Srinivasan and Mcdowell, 2009).

More realistic, robust and sub-field scale HSA (and CSA) delineations could be achieved by using a soil topographic index (STI) which integrates topographic data from Digital Elevation Models (DEMs) with maps of soil hydrological properties (e.g. Marjerison et al., 2011; Buchanan et al., 2013; Hahn et al., 2014). For example, the STI by Walter et al. (2002) is defined as $STI = \ln(^{\alpha}/_{t}an\beta K_sD) = \ln(^{\alpha}/_{t}an\beta) - \ln(K_sD)$. In this formulation, α (m) is the specific upslope drainage area per unit contour length, tanß is the surface slope gradient (radians), D is the local soil depth in metres to the restrictive layer (e.g. bedrock or fragipan), and K_s is the mean saturated hydraulic conductivity of the soil profile in metres per day above the restrictive layer. Areas with larger upslope drainage areas, shallower slopes, shallower soils and lower saturated hydraulic conductivities will have higher STI values (dimensionless), indicating higher runoff propensities.

Furthermore, high resolution DEMs derived from Light Detection and Ranging (LiDAR) allow HSA and CSA models to account for the effects of microtopographic features on surface runoff pathways and hydrological connectivity (Djodjic and Villa, 2015; Sharpley et al., 2011, 2015). For example, the HSA Index by Thomas et al. (2016) modifies the STI by reducing STI values in hydrologically disconnected drainage areas upslope of flow sinks such as depressions, pits or barriers that are topographically impeding overland flow. Thus high values are defined as HSAs (at high risk of runoff and surface P transport/delivery) rather than hydrologically active areas which (in terms of surface pathways) may not be hydrological connected to the stream and hence may not be at risk of delivering P.

Finally, the structure of CSA Indices may also need consideration. Most attempt to define one P loss risk score or load per field, by combining dissolved and particulate P losses from fertiliser P and legacy soil P sources via every transport pathway (Buczko and Kuchenbuch, 2007; Heckrath et al., 2008). However, it may be beneficial to develop specialised CSA Indices that focus on specific forms, sources, transport pathways and timings of P transfers (Djodjic and Bergström, 2005; Sharpley et al., 2015; Djodjic and Villa, 2015). Identifying specific types of CSAs, particularly those that are known to be locally dominant, would prioritise and minimise data collection, facilitate tailored recommendations of mitigation measures and best management practices, and aid model calibration and evaluation using specifically relevant empirical P loss data.

This study aimed to provide a 'next-generation' CSA Index that addresses these development needs. The study focused on the transfer risk of dissolved P from legacy soil P sources via surface runoff pathways during storm events, which is known to be one of the dominant pathways, forms and sources of P loss (Jordan et al., 2012; Sharpley et al., 2013b; Dodd et al., 2012; Mellander et al., 2015). The slow, persistent release of legacy P is recognised as a long term cause of water quality impairment and eutrophication, and a possible cause of the 'failure' of remediation strategies due to time lags between implementation and water quality response (Schulte et al., 2010; Wall et al., 2013; Sharpley et al., 2013b; Jarvie et al., 2013a,b; Withers et al., 2014). The objectives were to (1) use the soil WEP test to predict runoff dissolved P concentrations at the field scale based on relationships with empirical data, (2) use a HSA Index with high resolution LiDAR DEMs to predict steady-state runoff propensity and P transport/delivery potential, (3) combine the two factors to create a sub-field scale CSA Index map for onward targeting of mitigation measures and best management practices, and (4) evaluate CSA Index maps using measured P losses at catchment outlets.

2. Methods

2.1. Study sites and method overview

Four intensively monitored headwater agricultural catchments in Ireland, which are representative of intensive Irish agri-environmental conditions (Fealy et al., 2010), were selected for the study (Fig. 1). Catchments are described in detail elsewhere

Table 1
Characteristics of the four intensively monitored agricultural catchments used in the study.

	Arable A	Arable B	Grassland A	Grassland B
Area (ha)	1116	948	758	1207
Land use	Arable (54%) Grassland (39%)	Arable (33%) Grassland (49%)	Arable (6%) Grassland (84%)	Arable (20%) Grassland (77%)
Median slope (°)	3	3	4	3
Dominant soil drainage class	Well drained	Mixture of all classes	Well drained	Poorly drained, although well drained in uplands
Dominant soil types	Typical Brown Earths (88%), Gleyic Brown Earths (5%), Typical Groundwater Gleys (5%)	Stagnic Brown Earths (35%), Typical Surface-water Gleys (25%), Typical Brown Earths (22%)	Typical Brown Earths and Typical Brown Podzols (84%), Typical Surface-water Gleys (5%), Humic/ Typical Alluvial Gleys (4%)	Typical Surface-water Gleys or Groundwater Gleys (71%) Typical Brown Earths (29%)
Geology	Slate and siltstone	Calcareous greywacke and mudstone	Sandstone, mudstone and siltstone	Rhyolitic volcanic and slate
Dominant hydrological pathway following rainfall	Subsurface	Surface and subsurface	Subsurface	Surface and subsurface
Average annual rainfall (mm)[a]	1021	934	1117	1078
Average annual runoff (mm)[a]	548	444	618	520
Runoff coefficient[a]	0.54	0.48	0.55	0.48
Runoff flashiness (Q5/Q95)[b]	55	140	77	202
Average field size (ha)	3.32	2.70	2.00	3.04
Hedgerow density ($km^2\,km^{-2}$)	0.011	0.011	0.061	0.011
Ditch density ($km\,km^{-2}$)	1.3	2.3	1.7	5.7
Subsurface artificial drainage pipes (number observed)	34	Unknown	Unknown	40

[a] 1st Oct 2009–30 Sep 2014.
[b] Discharge measurements were ranked into percentile categories, from the 95th percentile low flows (Q95) to the 5th percentile high flows (Q5) (Jordan et al., 2012).

(e.g. Wall et al., 2011; Shore et al., 2013; Sherriff et al., 2015), and are summarised in Table 1. A method workflow for the study is summarised in Fig. 2.

2.2. Legacy soil P concentrations and mobilisation properties

Soil samples were collected across almost every field in each catchment (between 414 and 512) in two separate years, referred to hereafter as 'baseline' and 'resampled' years, and analysed for Morgan P, an agronomic soil P test which uses a buffered acetate-acetic acid reagent (Peech and English, 1944). These baseline and resampled years were Spring 2009 and Spring 2013 in Arable A and Grassland B, and Spring 2010 and Spring 2014 in Arable B and Grassland A. For each field unit (average size of 3 ha), 25 semi-random 10 cm deep soil samples were collected and homogenised according to national protocols (S.I. 31, 2014). For a small proportion of fields where data from one year were not collected, data from the other year were used.

To identify differences in P mobilisation potential, 27–35 spatially representative soil samples (10 cm depth) were also collected by Wall et al. (2012) in October 2011 in each catchment using a stratified grid (see Fig. 1), and analysed for soil P and mobilisation properties. These data included the agronomic Morgan P test, and the environmental WEP test which uses distilled deionized water (1:10 soil: solution ratio with 1 h shaking time) (Sharpley and Moyer, 2000). The Mehlich 3 test (M3; Mehlich, 1984) was also used to extract P, Al and Fe and determine the degree of P saturation (DPS) (P/(Al + Fe)) (Sims et al., 2002) to support the results. Relationships between these variables were then investigated using regression analysis and tested for significance using ANOVA. Land use specific relationships were also investigated, by differentiating soil samples into grassland or arable land use depending on the dominant land use within the last 5 years (records from Ireland's Department of Agriculture, Food and the Marine, pers.comm.).

Soil WEP concentrations (i.e. the mobile, soluble portion of legacy soil P that is immediately available for transport in runoff during rainfall events) were then estimated in each field using the catchment and land use specific relationships (regression line equations) found between Morgan P and WEP concentrations in the Wall et al. (2012) data. The field scale Morgan P and WEP concentrations were mapped in ArcGIS v10.0 and the field polygons rasterised to 2 m grid resolution. Distributions of concentrations were then analysed. Distributions of Morgan P concentrations were based on the Morgan P Index system of 1–4 within Ireland's EU Nitrates Directive (S.I. 378, 2006) regulations

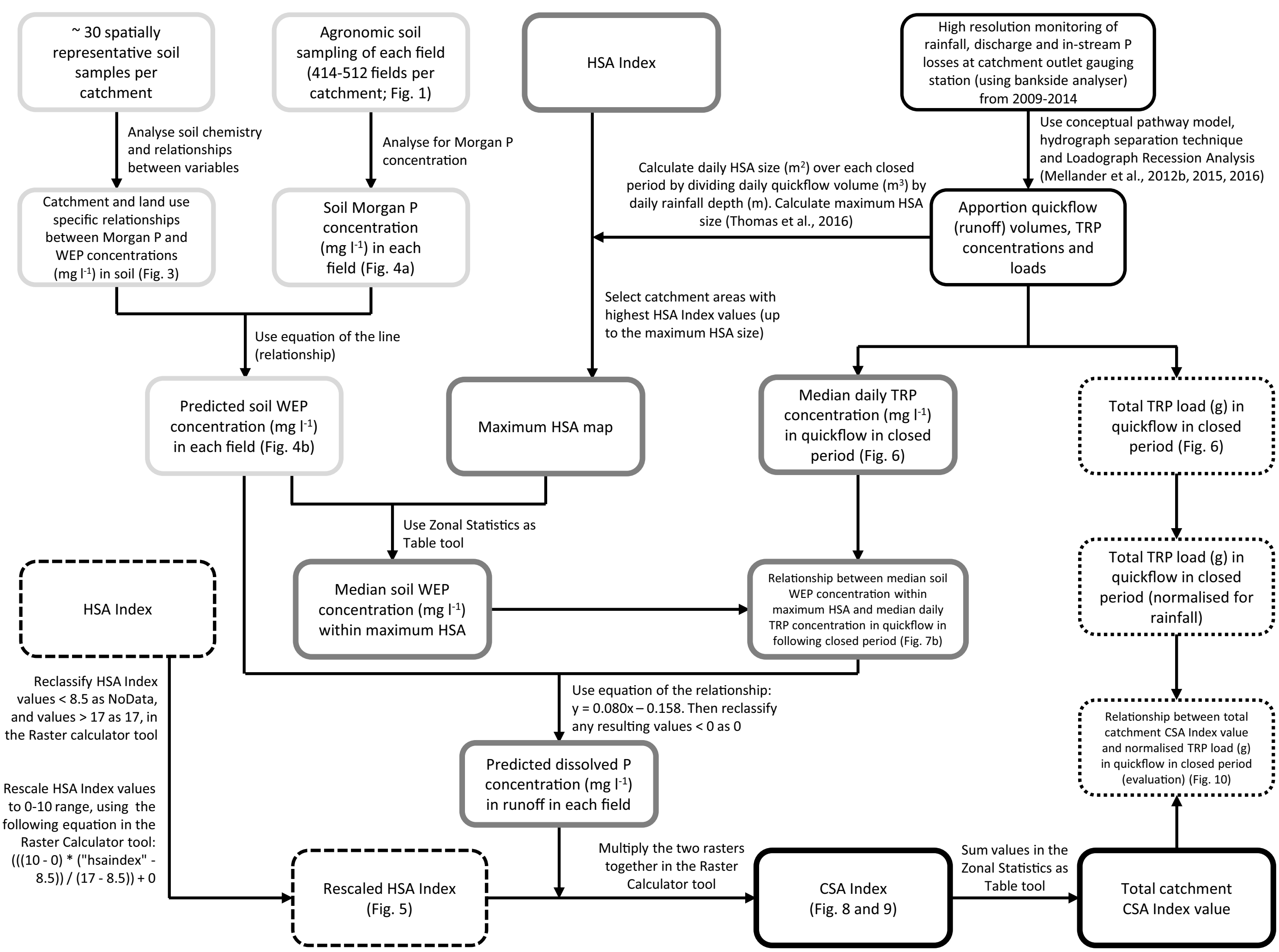

Fig. 2. CSA Index method workflow; determining soil WEP concentrations from agronomic soil samples (light grey), measuring P losses at the catchment outlet and apportioning losses from quickflow (runoff) (black), predicting dissolved P concentrations in runoff from soil WEP concentrations (dark grey), predicting runoff propensity using the HSA Index (dashed black), predicting relative differences in dissolved P loads in runoff per grid cell (CSA Index) and per catchment (total catchment CSA Index value) (thick black), and evaluating the CSA Index using measured TRP loads in quickflow at the catchment outlet (dotted black). ArcGIS tools used in the workflow are also indicated. P = phosphorus; WEP = water extractable P; TRP = total reactive P; HSA = hydrologically sensitive area; CSA = critical source area.

which are used to provide agronomic advice (i.e. Morgan P Index 3 and 4 soils are at and above agronomic optimum, respectively, whereas Morgan P Index 1 and 2 soils are deficient).

2.3. HSA Index

Runoff propensity was determined using the HSA Index by Thomas et al. (2016). First, 0.25 m grid resolution LiDAR DEMs (derived from an average of 38–46 bare-earth points m^{-2}) were acquired in each catchment (Supplementary Fig. 1) and resampled to 2 m resolution in ArcGIS. This resolution was deemed optimal for modelling HSAs in agricultural catchments dominated by microtopography (Thomas et al., 2017), as it captured flow-diverting features but also natural hillslope scale flow pathways, and removed high resolution topographic noise.

The DEMs were hydrologically corrected in SAGA GIS v2.1 to model fully connected flow pathways to the catchment outlet. This involved 'burning' a field-mapped open drainage channel network into the DEM, and identifying and filling flow sinks (pits and depressions) using the method by Wang and Liu (2006), which is designed for high resolution LiDAR datasets. Upslope drainage areas were then derived using the Deterministic Infinity (multiple flow direction) method by Tarboton (1997), and divided by the 2 m grid cell width to calculate α. Slope was computed from the un-hydrologically corrected DEM using the Zevenbergen and Thorne (1987) method, and used with α to derive a Topographic Wetness Index map based on the Beven and Kirkby (1979) equation-$\ln(^{\alpha}/_{t}an\beta)$. A $\ln(K_sD)$ raster was derived in ArcGIS using soils data from the Irish Soil Information System (Creamer et al., 2014) and additional soil surveys (R. Creamer, pers. comm.). STI maps (Supplementary Fig. 2) based on the equation by Walter et al. (2002) were then created by subtracting the $\ln(K_sD)$ raster from the Topographic Wetness Index using the raster calculator tool.

The HSA Index (2 m raster grid resolution) was then created by reducing STI values by 75% in upslope drainage areas of flow sinks that topographically impeded (hydrologically disconnected) overland flow as described in Thomas et al. (2016). This required the original 0.25 m LiDAR DEMs to define flow sinks, the resampled 2 m LiDAR DEMs to determine flow sink upslope drainage areas, and rainfall-quickflow measurements to determine HSA sizes and overland flow volumes.

HSAs were delineated by selecting the catchment area with the highest HSA Index values up to a threshold value. In this study, the threshold chosen was $\geq$8.5, which represented the maximum HSA size found in any of the four catchments (22.8% of Arable B) (Thomas et al., 2016). To ignore hydrologically insensitive areas where runoff and surface P transport does not occur, values <8.5 (which represent no runoff risk) were reclassified as NoData. Values >17 were typically found within the stream channel network or on roads, and were therefore reclassified as 17 to indicate equal, certain runoff propensity. HSA Index values were then rescaled between 0 and 10 (see Fig. 2).

2.4. Measuring runoff P concentrations and loads

To predict runoff P concentrations using soil WEP concentrations, and to evaluate the CSA Index using runoff P loads, in-stream total reactive P (TRP) concentrations were measured sub-hourly from 2009 to 2014 by bankside analysers (Hach-Lange Phosphax Sigma) at the catchment outlet gauging stations (Fig. 1). This fraction was deemed the most appropriate signal of P delivered from mobilised (soluble) legacy soil P sources. Mean hourly TRP loads were determined by integrating synchronous catchment outlet discharge measurements with TRP concentrations (Shore et al., 2014). A conceptual pathway model, hydrograph separation technique, and Loadograph Recession Analysis were then used to apportion TRP concentrations and loads from quickflow, interflow and slowflow pathways (Mellander et al., 2012, 2015, 2016). Only TRP concentrations and loads from quickflow pathways were used in the following analysis. Quickflow was assumed to be surface runoff only, although some preferential flow and tile/ditch drainage will also contribute.

As the CSA Index was designed to identify the spatial risk of legacy soil P transfers, only water quality data from the closed period for fertiliser applications in Ireland (15th October to 12th January) were selected, to remove potential noise from incidental P losses at other times of the year. The winter closed period also represents the most hydrologically active time of the year which experiences the highest quickflow (runoff) volumes and P transfers in the study catchments (Jordan et al., 2012) and in northwestern Europe (e.g. Ulén et al., 2011).

The relationship between the median soil Morgan P or WEP concentration within the maximum HSA extent (the catchment area which transports and delivers P) and the median daily TRP concentration in quickflow in the following closed period was investigated using regression analysis. The maximum HSA extent was delineated by selecting the catchment areas with the highest HSA Index values up to the known (catchment specific) maximum HSA size calculated by Thomas et al. (2016) using 2009–14 rainfall-quickflow data. As the relationship between soil WEP concentrations and quickflow TRP concentrations was strongest (see Section 3.3 and Fig. 7), the equation of the regression line was used to predict the dissolved P concentration in runoff (quickflow pathways) within each field in each catchment based on its soil WEP concentration, similar to approaches by Regan et al. (2010, 2014) and Vadas et al. (2005).

2.5. Developing the CSA Index and evaluating CSA maps

A CSA Index was created by multiplying the predicted runoff dissolved P concentration raster by the rescaled HSA Index (a steady-state proxy of runoff volume) in ArcGIS using the raster calculator tool. Thus higher CSA Index values indicate potentially higher dissolved P loads in runoff. CSAs were identified by selecting the catchment areas with the highest CSA Index values up to an arbitrary threshold value of $\geq$0.5, to identify high priority areas for mitigation and to demonstrate relative differences in CSA sizes between contrasting catchments.

CSA Index maps were then independently evaluated. First, the total CSA Index value was calculated for each catchment in baseline and resampled years using the zonal statistics tool in ArcGIS. Total catchment CSA Index values were then compared to the total TRP loads in quickflow in the following closed period using regression analysis. Due to the differences in rainfall between years and catchments, total quickflow TRP loads were first normalised by the mean total rainfall over a closed period (337 mm), assuming a linear relationship. Total catchment CSA Index values were used for evaluation rather than the proportion of the catchment identified as a CSA (using the CSA Index threshold value), as the latter approach ignores potentially large HSAs with lower CSA Index values that are also contributing to overall P losses.

3. Results

3.1. Soil P concentrations and mobilisation potential

Mean Morgan P, WEP, DPS and ancillary variables of the soil samples by Wall et al. (2012) are shown in Table 2. Samples are also differentiated into grassland or arable soils. Relationships between key variables (Morgan P, WEP and DPS) are shown in Fig. 3. Results show strong positive relationships between Morgan P and DPS concentrations in all catchments (Fig. 3c), including when samples were differentiated between land use.

Moderate-to-strong positive relationships were found between DPS and WEP concentrations in all catchments (Fig. 3a). Grassland B had significantly higher WEP concentrations for a given DPS value compared to Arable A ($P < 0.05$), and Grassland A and Arable B had similar relationships (Wall et al., 2012). Also, within each catchment, grassland soils had significantly higher WEP concentrations for a given DPS value compared to arable soils, described by significantly higher intercepts ($P < 0.001$) rather than differences in slope (insignificant). Comparisons between land uses in Grassland A could not be undertaken due to a lack of arable field samples.

Moderate-to-strong positive relationships were also found between Morgan P and WEP concentrations in all catchments. Arable A had significantly lower WEP concentrations for a given Morgan P concentration compared to Arable B ($P < 0.05$), Grassland A ($P < 0.05$) and Grassland B ($P < 0.001$) (Fig. 3b). Also, within each catchment, grassland soils had significantly higher WEP concentrations for a given Morgan P concentration compared to arable soils, described by significantly higher intercepts ($P < 0.001$) rather than differences in slope.

Morgan P concentration maps for each catchment are shown for the baseline year in Fig. 4a, based on the field scale soil sampling. WEP concentration maps are also shown in Fig. 4b, estimated using the catchment and land use specific relationships found between Morgan P and WEP concentrations in the Wall et al. (2012) data (b). Distributions of Morgan P Index and WEP concentrations are shown for both baseline and resampled years in Fig. 4c and d respectively. All catchments (particularly Grassland A and Arable A and B) had fields with excessive Morgan P concentrations above agronomic optimums, indicating legacy soil P source pressures (Wall et al., 2012). However, WEP concentrations were lowest in Arable A, with Grassland B indicating higher soil P mobility, followed by Grassland A and Arable B. All catchments showed variations in Morgan P and WEP concentrations between baseline and resampled years, with all catchments except Arable B showing decreases in the highest concentrations.

3.2. HSA Index maps

The HSA Index maps for each catchment are shown in Fig. 5a (rescaled from HSA Index maps in Thomas et al., 2016). Higher values in red indicate higher hydrological sensitivity to rainfall and higher runoff propensity, whereas lower values in green indicate lower runoff propensity. Catchment areas without HSA Index values are deemed hydrologically insensitive to rainfall even during the largest storm events (in terms of runoff generation), and are therefore not considered within CSA Index calculations. As described by HSA Index value distributions in Fig. 5b and in Thomas et al. (2016), Grassland B has the highest runoff propensity, followed by Arable B, Arable A and Grassland A.

3.3. TRP concentrations and loads in quickflow, and relationships with soil Morgan P and WEP concentrations

Median daily TRP concentrations and total TRP loads in quickflow pathways during 2009–10 to 2014–15 closed periods

Table 2
Mean Morgan P, WEP, M3-P, M3-Al and M3-Fe concentrations, pH, %OM and DPS of soil samples collected and analysed by Wall et al. (2012). Samples are also differentiated into grassland and arable soils using the average land use of the previous five years.

		Arable A	Arable B	Grassland A	Grassland B	All catchments
n	All	35	29	27	30	121
	Arable	24	12	3	7	46
	Grassland	11	17	24	23	75
Morgan P[a] (mg l^{-1})	All	7.8	7.1	6.6	5.0	6.7
	Arable	8.4	6.9	5.9	4.3	7.2
	Grassland	6.6	7.2	6.7	5.2	6.3
WEP[b] (mg l^{-1})	All	2.8	3.9	4.0	4.0	3.6
	Arable	2.0	2.4	3.0	2.1	2.2
	Grassland	4.5	5.0	4.1	4.6	4.5
M3[c]-P (mg kg^{-1})	All	79.2	64.7	76.4	47.0	67.1
	Arable	82.5	75.2	80.4	55.3	76.3
	Grassland	72.1	57.3	75.9	44.4	61.5
M3-Al[d] (mg kg^{-1})	All	1115	955	874	908	971
	Arable	1147	1039	948	1070	1094
	Grassland	1044	896	864	858	896
M3-Fe[e] (mg kg^{-1})	All	249	354	484	373	357
	Arable	242	325	460	316	289
	Grassland	263	375	487	391	399
M3-Al + Fe (mg kg^{-1})	All	1363	1310	1357	1281	1329
	Arable	1389	1364	1408	1386	1383
	Grassland	1307	1271	1351	1249	1295
pH	All	6.4	5.7	5.9	5.9	6.0
	Arable	6.5	5.8	5.7	5.9	6.2
	Grassland	6.2	5.7	5.9	5.9	5.9
OM[f] (%)	All	7.8	6.5	7.5	7.6	7.4
	Arable	6.1	4.8	5.7	5.6	5.6
	Grassland	11.5	7.7	7.7	8.2	8.4
DPS[g] (%)	All	18.0	16.6	19.1	13.7	16.8
	Arable	18.3	17.5	18.9	14.0	17.5
	Grassland	17.3	16.0	19.2	13.6	16.5

[a] Phosphorus.
[b] Water extractable P.
[c] Mehlich 3 test (Mehlich, 1984).
[d] Aluminium.
[e] Iron.
[f] Organic matter.
[g] Degree of P saturation (P/(Al + Fe)).

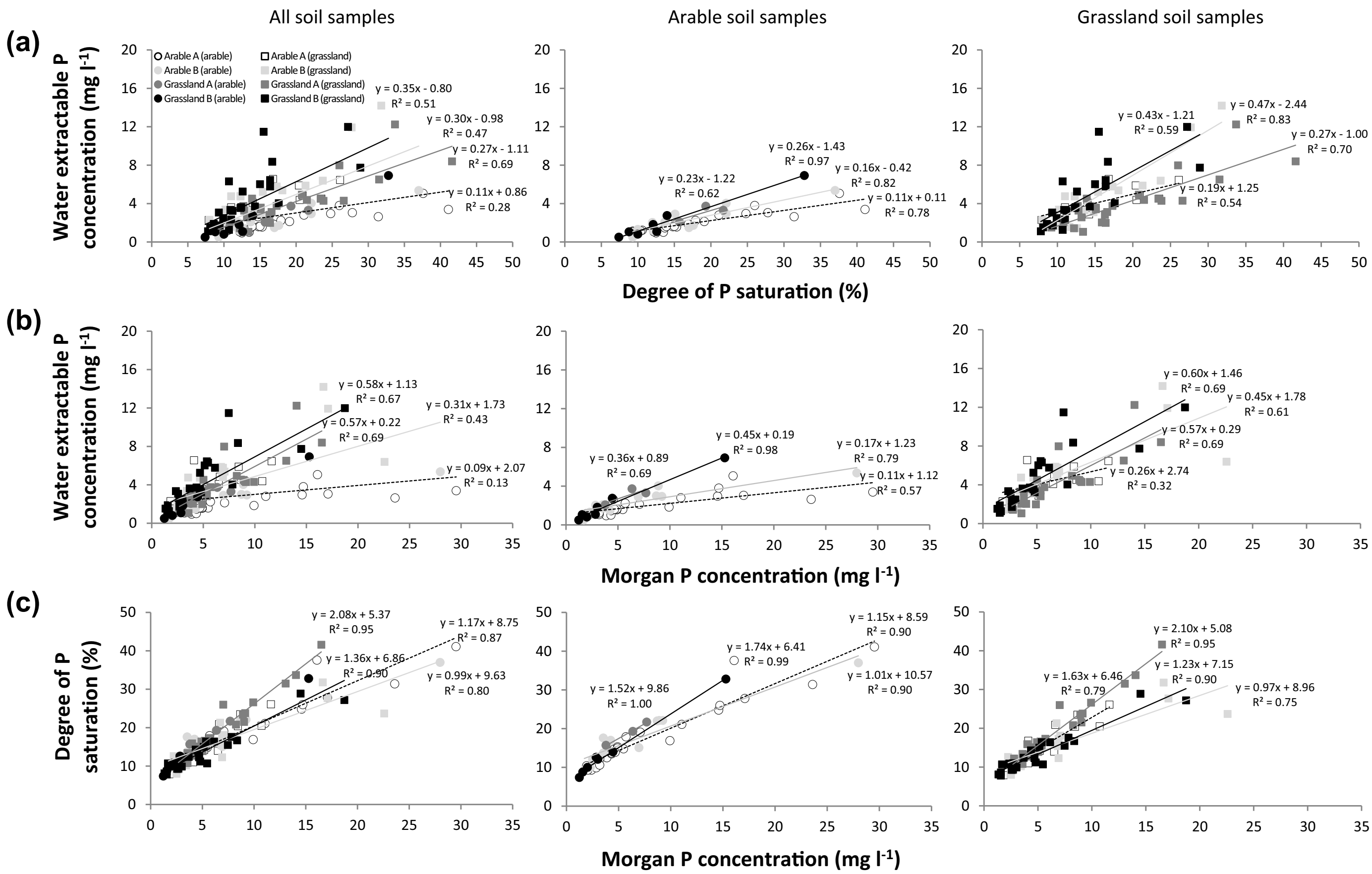

Fig. 3. Relationships between (a) %DPS (P/(Al + Fe)) and WEP concentration, (b) Morgan P and WEP concentration, and (c) Morgan P concentration and %DPS, for soil samples collected by Wall et al. (2012). Samples are differentiated by catchments (left), and also by arable (middle) and grassland (right) land uses. P = phosphorus; WEP = water extractable P; DPS = degree of P saturation.

are shown in Fig. 6. General trends showed that Grassland A experienced the highest TRP concentrations in quickflow (runoff), followed by Arable B, Grassland B and Arable A. Although concentrations fluctuated between years within each catchment, trends were relatively small, except for a large reduction in Grassland A during the 2013–14 closed period. Contrastingly, Grassland B consistently experienced the highest total TRP loads in quickflow pathways between 2009–10 to 2013–14, compared to Grassland A which had the second lowest loads. Variations in total TRP loads between years were similar across each catchment, although Grassland B showed much larger reductions between 2009–10 and 2010–11.

The relationship between the median Morgan P or WEP concentration ($mg\,l^{-1}$) in the soil within the maximum HSA and the median daily TRP concentration ($mg\,l^{-1}$) in quickflow in the following closed period from when the soil was sampled is shown in Fig. 7. Arable B and Grassland A resampled years were not included due to the unavailability of quickflow data in the following closed period (2014–15). Although a strong positive relationship was indicated between Morgan P concentration and TRP concentration in quickflow in three of the four catchments, Arable A had relatively high Morgan P concentrations but very low quickflow TRP concentrations, and hence the overall relationship was weak. Conversely, median WEP concentrations had a strong positive correlation with TRP concentrations in quickflow across all catchments ($r^2 = 0.73$). Hence, WEP was shown to be a stronger indicator of dissolved P concentrations in runoff within each field unit. Predicted dissolved P concentrations in runoff $<0\,mg\,l^{-1}$ (an artefact of the equation intercept) were reclassified as $0\,mg\,l^{-1}$.

3.4. CSA Index maps and evaluation

CSA Index maps and value distributions are shown in Fig. 8 for baseline and resampled years. CSA Index values typically ranged between 0.1 and 2. A close-up view is shown in Fig. 9, which also indicates breakthrough and delivery points where P is potentially transported between fields and delivered to the drainage network, respectively. CSAs (catchment areas with the highest CSA Index values) are clearly identifiable at the sub-field scale, as well as individual CSA (runoff) pathways. Using an arbitrary CSA Index threshold value of ≥ 0.5 to delineate CSAs, Grassland B had the largest CSAs (5.6% and 4.1% of the catchment area in baseline and resampled years, respectively), followed by Arable B (2.9% and 3.0%), Grassland A (2.9% and 2.4%) and Arable A (1.4% and 1.1%).

The relationship between the total catchment CSA Index value and the total TRP load within quickflow in the following closed period is shown in Fig. 10. The strong relationship ($r^2 = 0.86$) indicates that the CSA Index can accurately predict relative differences in dissolved P loads in runoff between contrasting catchments and land management. Grassland B had proportionately the largest CSAs because it had the greatest P transport potential (HSA Index) which coincided with large mobile P source pressures (WEP). Arable A had the smallest CSAs as it had the lowest runoff risk and the lowest WEP concentrations. Although Grassland A had large WEP concentrations, it had the second lowest P transport potential and hence a low coincidence of mobile P sources and HSAs. Arable B was at second highest risk of dissolved P transfers because it had large mobile P sources coinciding with relatively large HSAs.

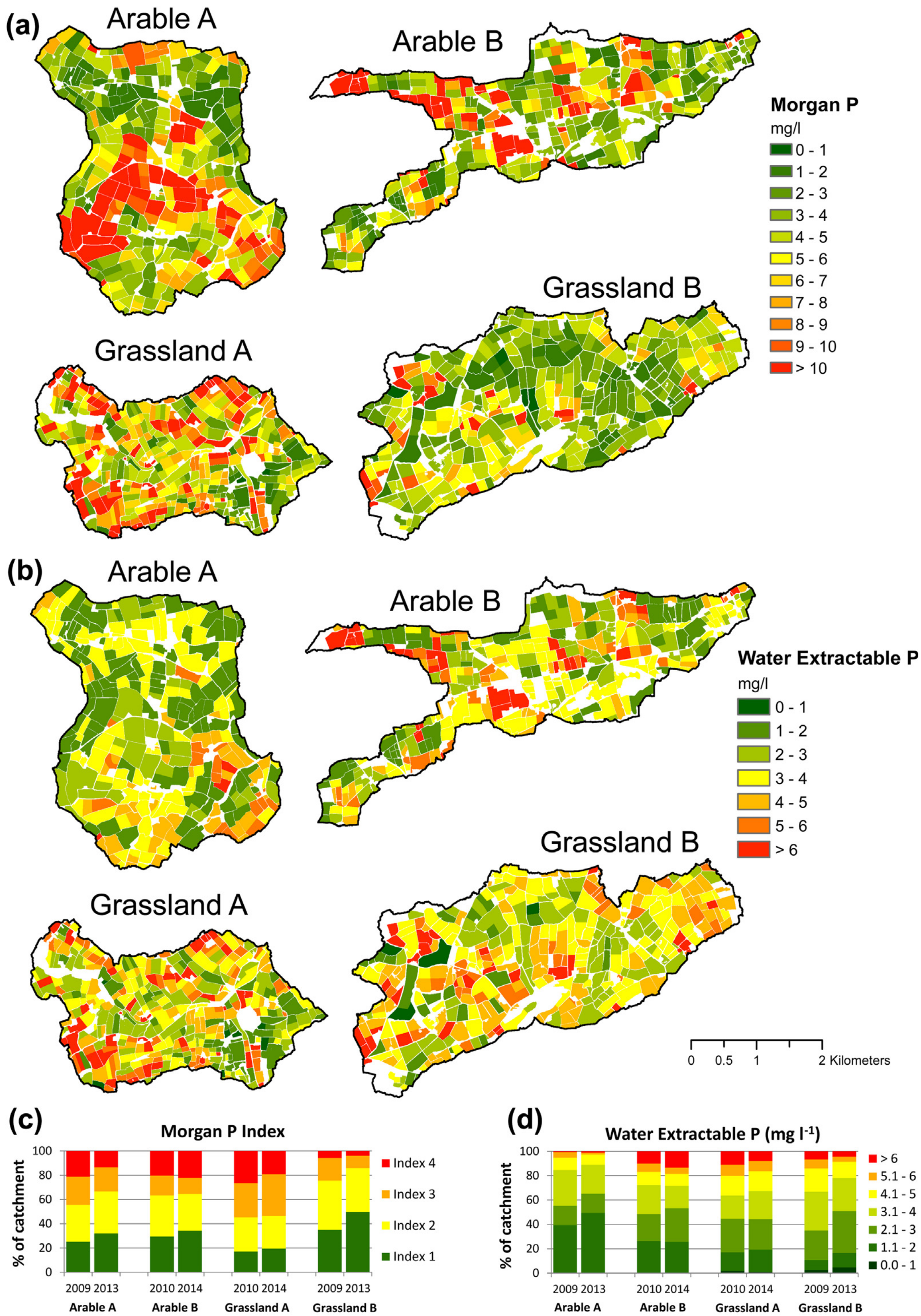

Fig. 4. Maps of (a) Morgan P concentrations (mg l^{-1}) and (b) WEP concentrations (mg l^{-1}) for the baseline year for each field unit. Also shown are the proportion of the catchment area in each Morgan's P Index 1–4 (c) and WEP concentration (d) for baseline and resampled years. WEP concentrations were predicted using the catchment and land use specific relationships with Morgan P concentrations found in Fig. 3b. P = phosphorus; WEP = water extractable P.

4. Discussion

4.1. The importance of mobilisation potential of legacy P

The moderate-to-strong positive relationships found between DPS and WEP concentrations within all catchments (Fig. 3a) agrees with other studies which show that different soil types have different capacities to chemically retain and attenuate P (i.e. P sorption sites from Al and Fe) and hence have different potentials for P mobilisation and losses (e.g. Maguire and Sims, 2002; Sims et al., 2002; Daly et al., 2001). The relationships between DPS and WEP concentrations were similar to the relationships between Morgan P and WEP (Fig. 3b). This corresponds with findings from Fig. 3c and other studies which show that agronomic soil P tests relate well to DPS (Kleinman et al., 1999; Kleinman and Sharpley, 2002; Pautler and Sims, 2000).

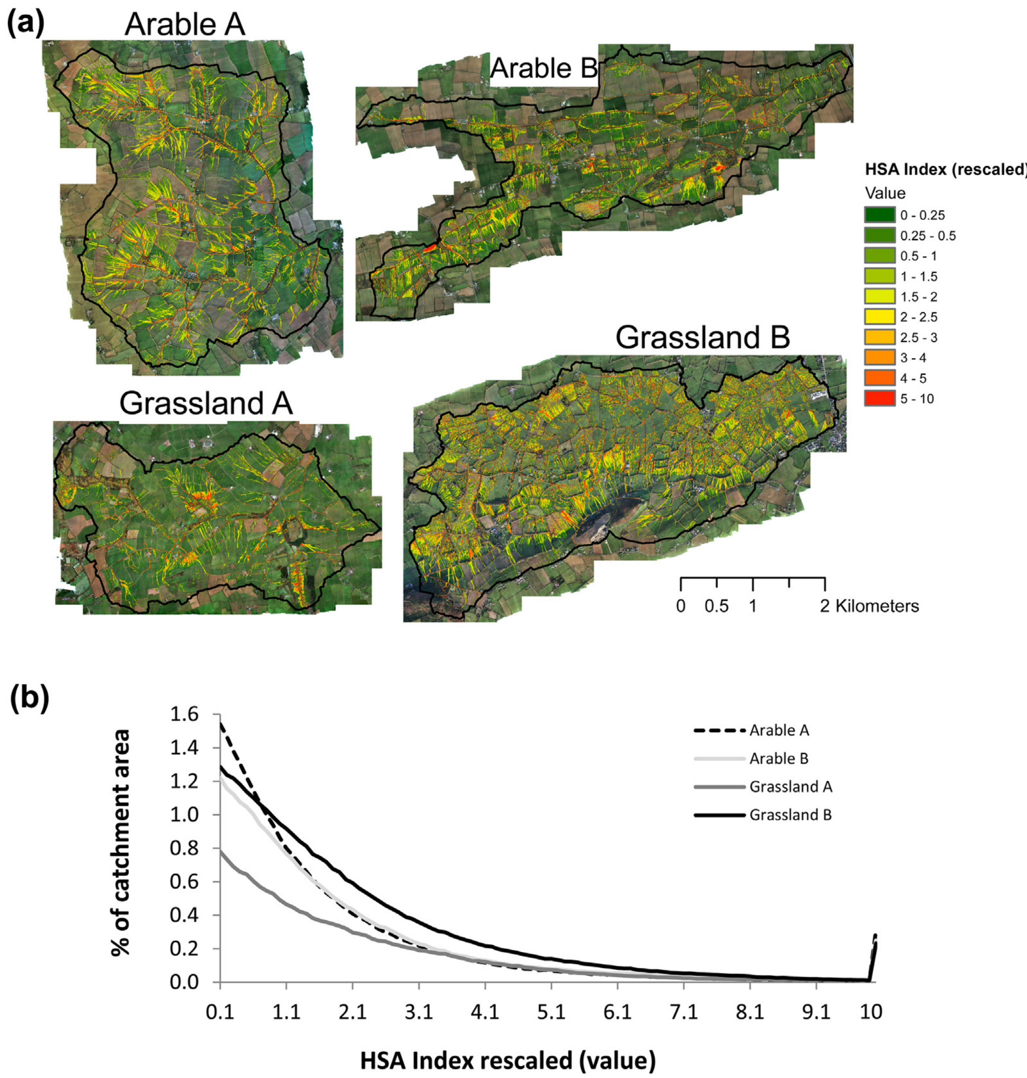

Fig. 5. (a) Maps of the rescaled HSA Index, overlaying orthophotos. Higher values in red and lower values in green indicate higher and lower runoff propensity, respectively. (b) Rescaled HSA Index value distributions. HSA = hydrologically sensitive area.

However, the relationship between DPS or Morgan P and WEP concentrations was found to be both catchment and land use specific (Fig. 3a,b; Wall et al., 2012). For example, although Grassland B had the lowest mean Morgan P concentration and DPS in the Wall et al. (2012) data, it had the highest mean WEP concentration (Table 2). Conversely, Arable A showed relatively high Morgan P and DPS, but the lowest WEP concentrations. This is explained both by natural differences in soil chemistry, and man-made differences due to fertiliser/lime applications or the vertical stratification of P and organic matter in grassland soils (see Table 2) which create fewer and weaker bonds compared to mineral-dominated sublayers where P is incorporated in tillage soils (Daly et al., 2001; Ulén et al., 2011; Daly and Casey, 2005; Page et al., 2005; Torbert et al., 2002; Hooda et al., 2001). Furthermore, change points and plateaus have sometimes been observed in the relationships between DPS, Morgan P and WEP (Daly et al., 2015; Sharpley et al., 2004; McDowell and Sharpley, 2001). It is therefore likely that, when applying the CSA Index elsewhere, the nature of the relationships between variables found in this study may differ.

4.2. The dominance of P transport (HSAs) in CSAs

Results indicate that P transport (via HSAs) is a more dominant factor of dissolved P CSAs than mobile soil P sources (WEP concentrations). Grassland A had the highest average TRP concentrations in quickflow (runoff) in the closed period, followed by Arable B, Grassland B and Arable A (Fig. 6). However, it was Grassland B that had the highest quickflow TRP loads, followed by Arable B, Grassland A and Arable A (Fig. 6), indicating that runoff volumes from HSAs were more important in driving the delivery of soil WEP to the watercourse than the WEP concentration. These results are supported by similar international findings in a range of catchments where P transfers are predominantly via surface pathways (e.g. Buda et al., 2009; Campbell et al., 2015; Kyllmar et al., 2006; Bergström et al., 2007). This confirms the P transfer continuum concept whereby mobilised P cannot be delivered unless it is transported via hydrologically connected pathways (Haygarth et al., 2005; Thomas et al., 2016). However, it is clear that the CSA Index requires both factors to accurately predict spatial dissolved P loss risk, as differences in soil WEP concentrations

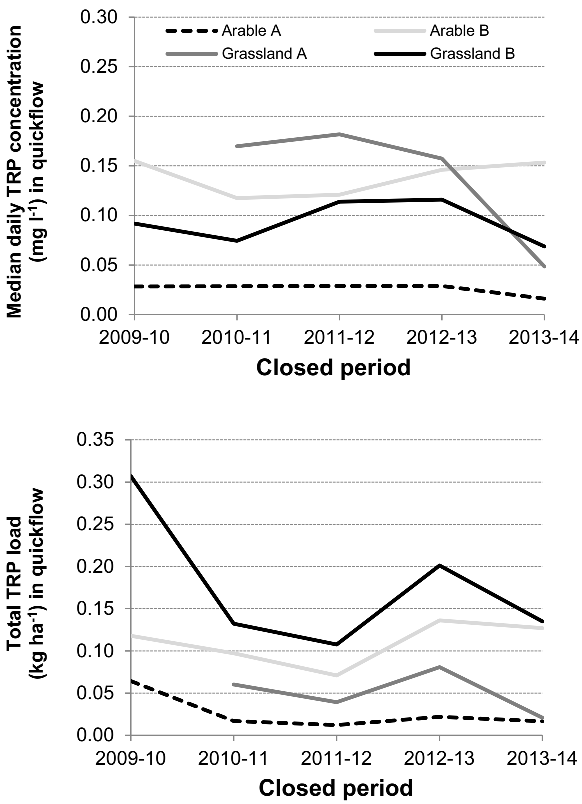

Fig. 6. Median daily TRP concentrations and total TRP loads in quickflow in 2009–10 to 2014–15 closed periods for fertiliser applications (15th October–12th January). TRP = total reactive phosphorus.

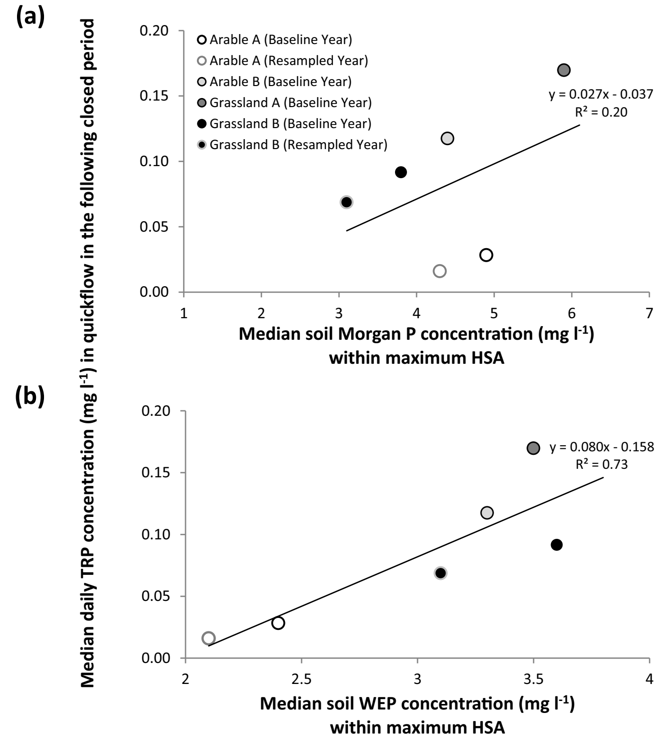

Fig. 7. Relationship between (a) median Morgan P concentration (mg l^{-1}) or (b) median WEP concentration (mg l^{-1}) in the soil within the maximum HSA extent (calculated in Thomas et al., 2016) and the median daily TRP concentration (mg l^{-1}) in quickflow in the following closed period. Arable B and Grassland A resampled years were not included as quickflow data was unavailable in the following closed period (2014–15). P = phosphorus; WEP = water extractable P; TRP = total reactive P.

between baseline and resampled years caused considerable changes in CSA sizes and total catchment CSA Index values.

4.3. Implications and recommendations for agricultural policy, farming practices and mitigation

The findings from this study (Figs. 3 and 7), in combination with others mentioned previously, suggest that the soil WEP test is better at predicting runoff P concentrations over a diverse range of catchments compared to an agronomic soil P test (such as Morgan P), and should be used to indicate environmental P loss risk. Soil WEP should ideally be measured directly (which could further improve predictions), or at least inferred using catchment and land use specific relationships with Morgan P as demonstrated in this study.

The dissolved P CSA Index was able to identify CSAs at the sub-field scale (Figs. 8 and 9) as well as individual CSA pathways of P losses and drainage channels at higher risk of delivery. This allows field and sub-field scale mitigation measures and best management practices to be implemented at precisely targeted locations, to improve cost-effectiveness and minimise disturbance to farmers (Doody et al., 2012; Buda et al., 2012; Sharpley et al., 2011). As the CSA Index uses the HSA Index, only CSAs that are hydrologically connected to the watercourse are identified. This further improves the targeting and effectiveness of measures and avoids unnecessary implementation at hydrologically disconnected pathways.

A range of CSA measures are available as part of a 'treatment-train' approach to mitigation (Ferrier et al., 2005; Campbell et al., 2004) that target different stages of the source-mobilisation-transport-delivery-impact continuum. For example, targeted nutrient management can reduce soil P sources at CSAs (Murphy et al., 2015), and chemical amendments of soils (and fertilisers/manures) such as lime, gypsum or alum additions can reduce P mobility (Fenton et al., 2011; Murphy and Stevens, 2010; Murphy and Sims, 2012; Smith et al., 2001). Measures that focus on reducing P transport by slowing, impeding or treating runoff from HSAs, such as runoff attenuating features, riparian buffer strips, wetlands, permeable reactive interceptors and modifying ditches (Schoumans et al., 2014; Wilkinson et al., 2014; Ockenden et al., 2014; Fenton et al., 2014; Shore et al., 2015) can also be targeted at vulnerable 'breakthrough points' at field boundaries and 'delivery points' at the watercourse (Fig. 9) (Thomas et al., 2016, 2017).

Compared to using an agronomic soil P test alone, the dissolved P CSA Index may lessen the burden on farmers in terms of the need to decline certain high legacy soil P stores (Schulte et al., 2010; Wall et al., 2013; Sharpley et al., 2012). For example, current regulations that constrain soil P amendments for water quality protection prohibit the application of inorganic fertiliser P on fields with soil P status above agronomic optimum (Morgan P Index 4 in Ireland) (Humphreys, 2008). However, although 4.0–26.5% of catchment areas were Morgan P Index 4 soils (Fig. 4a,c), only 1.1–5.6% of catchment areas in this study were predicted to be CSAs based on the threshold value used (Fig. 8), and specific locations differed. This follows findings by Thomas et al. (2016) that only 2.9–8.5% and 6.2–22.8% of catchment areas were HSAs during upper quartile and maximum storm events respectively. In many fields, high soil test P (Morgan P) coincided with relatively low P mobility (soil WEP concentrations) and/or low runoff potential (HSA Index values), and hence were not CSAs. Conversely, some fields with lower soil test P had relatively high P mobility and/or high runoff potential, and hence were considered to be CSAs. Therefore P application restrictions could be reconsidered in non-CSAs, for example to relieve manure storage pressures over winter, although care would

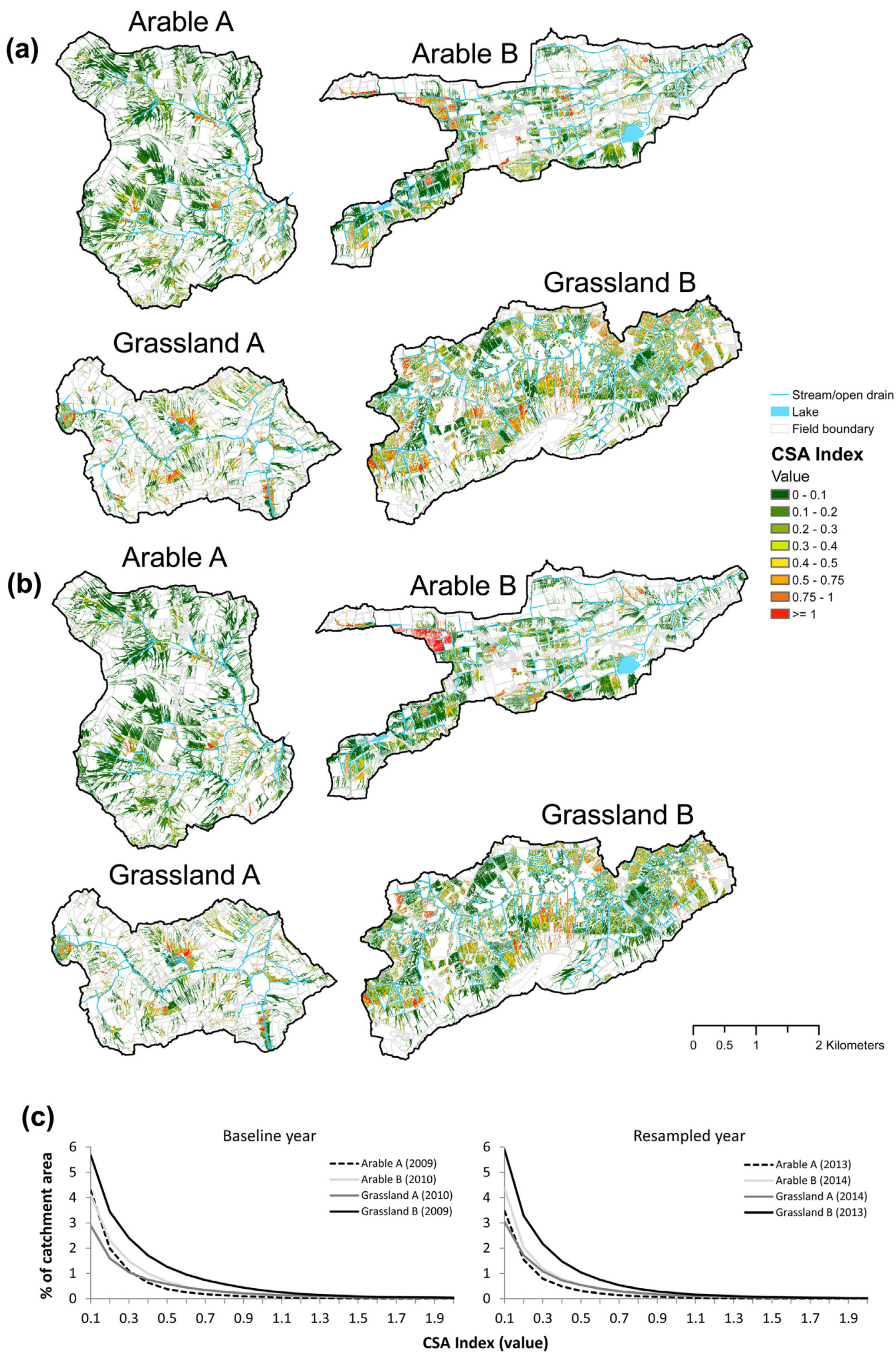

Fig. 8. CSA Index maps for (a) baseline and (b) resampled years, and (c) the proportion of the catchment area with each CSA Index value. Higher (red) and lower (green) CSA Index values indicate higher and lower predicted dissolved P loads in runoff, respectively. CSAs are identified as the catchment areas with the highest CSA Index values (e.g. $\geq$0.5). Catchment areas without CSA Index values are not hydrologically sensitive to rainfall according to the HSA Index, and hence are not considered in CSA Index calculations. CSA = critical source area; HSA = hydrologically sensitive area.

be needed to avoid potential increases in subsurface and groundwater P transfers in the long term (following time lags).

Next-generation screening tools are now also available that characterise and/or monitor catchments and sub-catchments to identify those at higher risk of P transfers (e.g. Packham et al., 2014; Brazier et al., 2005). These should be used to target and prioritise implementation of the finer scale HSA and CSA Indices presented here.

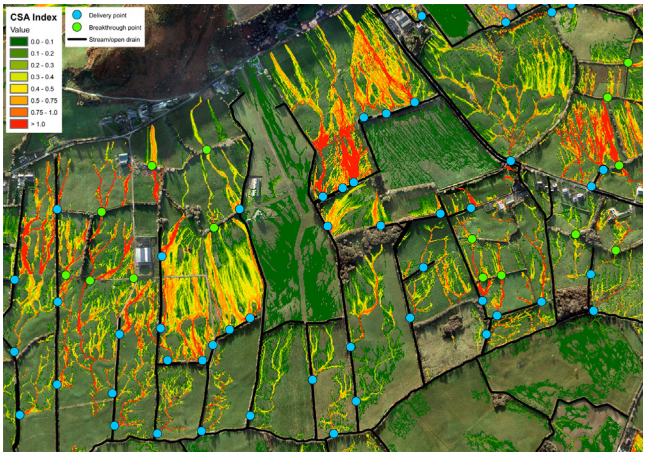

Fig. 9. Close-up view of a CSA Index map. Also indicated are breakthrough points (in green) and delivery points (in blue) where CSA pathways cross field boundaries and are delivered to the drainage network, respectively. These are where sub-field scale mitigation measures could be targeted and prioritised. CSA = critical source area.

4.4. Other considerations

Further work is required to evaluate the CSA Index by demonstrating (through short and long-term monitoring) that targeted mitigation measures and best management practices at the identified CSAs causes in-stream declines in P loads and associated improvements in water quality (Doody et al., 2012; Sharpley et al., 2009). This is complicated by the fact that water quality and ecological responses to reduced P loadings differ between rivers and catchments, as recovery trajectories can be decoupled, non-linear and characterised by thresholds, alternative stable-states and time lags (Jarvie et al., 2013b). The CSA Index threshold value used to delineate CSAs may need refining if mitigation of those areas is found to be insufficient at counter-acting eutrophication, for example by using the relationship in Fig. 10 in combination with environmental thresholds. The tool also needs to be evaluated in terms of cost-benefits and farm economic viability and compared to that of no intervention or intervention at excessively high (surplus) P soils or HSAs alone (Kronvang et al., 2009; Ghebremichael et al., 2013).

Limitations should be acknowledged in the approach which may account, for example, for the (albeit small) unexplained variability in Fig. 7b and Fig. 10. As the accuracy of the HSA Index depends in part on the accuracy of soil series maps, additional soil sampling and/or expert judgement may be required if national scale (1:250,000) datasets are used. Also, the HSA Index method requires the estimation of the HSA size to determine flow sink 'fill and spill' potential. In ungauged catchments, the HSA size found in Thomas et al. (2016) in the catchment with the most similar STI value distribution could be used. Alternatively, local rainfall-runoff curve numbers could be applied, or approaches outlined by Hrachowitz et al. (2013). Local knowledge of soil compaction from poaching and trafficking, which can cause infiltration-excess overland flow, should be accounted for in the K_sD component of the HSA Index. Furthermore, in fields with subsurface artificial drainage, K_s will increase, but this was not considered in the HSA Index as their locations, design, age, depth and effectiveness are unknown. Additionally, the steady-state CSA Index does not account for differences in rainfall and hence hydrological activity of HSAs between catchments, regions and years. Finally, erroneous flow pathways and flow sinks may occur as artefacts of DEM vertical error.

5. Conclusions

Conventional CSA definitions of P transfers have a number of limitations which need to be addressed using the latest scientific and technological advances if mitigation measures and best management practices are to be cost-effectively targeted. A new 'next generation' CSA Index of dissolved P losses from legacy soil P is presented here that addresses these concerns. Specifically, the GIS-based approach uses soil WEP concentrations, rather than agronomic soil P, to predict mobile soil P sources. It also uses a new soil-topographic HSA Index derived from high resolution (0.25 m and 2 m) LiDAR DEMs, rather than watercourse proximity, to realistically define runoff propensity, P transport potential and hydrological connectivity. The CSA Index was applied to four intensively monitored agricultural catchments representing different agri-environmental conditions. Sub-field scale CSA maps were generated and evaluated against total TRP loads in quickflow in the 2009–2014 winter closed periods ($r^2 = 0.86$).

In conclusion:

- Relationships between soil Morgan P, WEP and DPS were both catchment and land use specific, with significantly higher WEP release in grassland soils compared to arable soils. WEP concentrations should ideally be measured directly, or at least

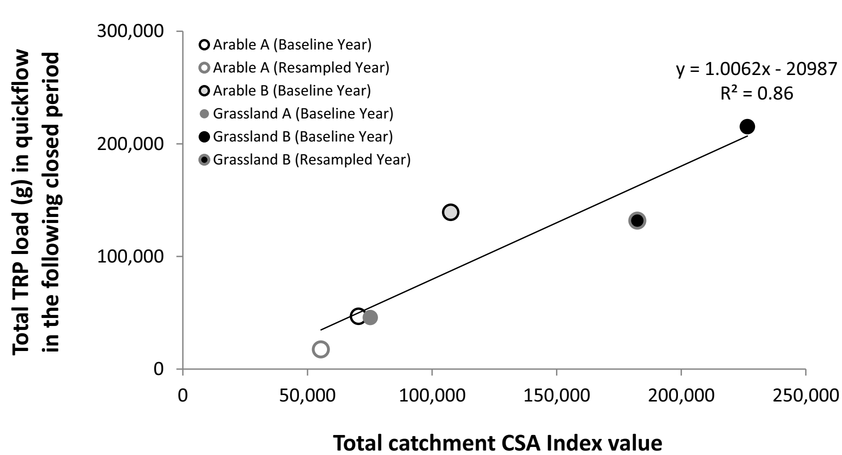

Fig. 10. Relationship between the total CSA Index value for each catchment in baseline and resampled years and the total TRP load (g) in quickflow in the following closed period. Due to the differences in rainfall between years and catchments, loads were normalised by the mean total rainfall over a closed period (337 mm). Arable B and Grassland A resampled years were not included as quickflow data was unavailable in the following closed period (2014–15). CSA = critical source area; TRP = total reactive phosphorus.

inferred using catchment and land use specific relationships with Morgan P or DPS

- Hydrological transport (within HSAs) was a more important factor for dissolved P transfers than soil P mobilisation. Thus mitigation strategies and policies should first prioritise impeding P transport in surface runoff
- The proportion of catchment areas defined as high risk to water quality according to above agronomic optimum soil P alone (the current risk assessment used in the study catchments) was between 4.0–26.5%, compared to a CSA size of 1.1–5.6% delineated using the fully developed CSA Index. This provides an important selling point for the farming community by highlighting locations where current P application restrictions could be reconsidered
- Targeting mitigation measures at breakthrough points and delivery points where CSA pathways cross field boundaries and are delivered to the drainage network, respectively, could further reduce the potential area of intervention and improve cost-effectiveness.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.agee.2016.09.012.

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